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TITLE: DEPOLARIZATION OF DIFFUSING SPINS BY PARAMAGNETIC IMPURITIES

MASTER

AUTHOR(S): M. E. Schillaci, R. L. Hutson, R. H. Heffner, and M. Leon

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University of California

LOS ALAMOS SCIENTIFIC LABORATORY
Post Office Box 1663 Los Alamos, New Mexico 87545
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DEPOLARIZATION OF DIFFUSING SPINS BY PARAMAGNETIC IMPURITIES*

M. E. Schillaci, R. L. Hutson, R. H. Heffner, and M. Leon
Los Alamos Scientific Laboratory, Los Alamos, NM 87545 USA

S. A. Dodds and T. L. Estle
Rice University, Houston, Texas 77001 USA

We study the depolarization of diffusing spins (muons) interacting with dilute paramagnetic impurities in a solid using a simple computational model which properly treats the muon motion and preserves correct muon-impurity distances. Long-range (dipolar) and nearest-neighbor (contact) interactions are treated together. Diffusion parameters are deduced and model comparisons made for AuGd (300 ppm).

An interesting problem for μSR research is to understand the diffusive behavior of muons in metals. In a recent series of experiments, we approached this problem by studying μ^+ depolarization in target materials that have negligibly small nuclear dipole moments and are doped with small concentrations of paramagnetic impurities. In this way, we can extend the range of hosts and temperatures where it is possible to determine muon diffusion coefficients. In addition, we can gain information about the impurity spin dynamics which is not accessible to conventional techniques, as well as determine the nature of the muon-impurity interaction. A full description of the experiments is given in another paper at this conference[1]; we wish to report on a computational model which properly treats the muon motion and preserves correct muon-impurity distances, while accounting for impurity spin fluctuation and some muon-impurity interactions.

Much of our data has been analyzed using the model of F.M. Richards.[2] One form of this model is based on the steady-state solution of the diffusion equation with depolarization and is designed for treating long-range interactions, such as dipole-dipole. The lattice, and therefore the muon motion, is assumed to be continuous. In another form of the model, the muon is assumed to execute a random walk (self-avoiding) in discrete hops through the lattice. This form is more suitable for treating nearest-neighbor interactions, such as a contact interaction. Difficulties that arise in applying this model include:

(1) Because the depolarization term is proportional to r^{-6} for a dipolar interaction, the results are sensitive to the discrete distances accessible to the muon when close to the impurity. Thus, the assumption of a continuous lattice is not a good one at small distances.

(2) The random walk of the muon is not actually self-avoiding, as assumed.

(3) Both long-range and nearest-neighbor interactions may be present and should be treated together.

One way to avoid these difficulties is to treat the muon motion exactly with a Monte Carlo simulation code. However, when dealing with dilute impurity concentrations of the order of 100 ppm atomic, this technique consumes a great deal of computer time. The approach described here preserves some of the calculational simplicity of the diffusion model approach while avoiding both the difficulties mentioned above as well as a lengthy Monte Carlo calculation. We proceed with a description of our model.

First, we enumerate all possible muon locations in a specified volume around an impurity and list them according to distance from the impurity, specifying the number of sites at each radius. The muon sites thus constitute shells surrounding the impurity. Initially, the polarization residing on each shell is proportional to the number of sites on each shell, i.e., uniform polarization. We assume the muon must go to a nearest-neighbor site each time it hops. The transition probability per hop between shells can then be determined. Polarization is exchanged among shells according to these probabilities and lost according to whatever depolarization mechanism is being considered.

We take AuGd (300 ppm) as an example to illustrate the model. Gold has an FCC lattice ($a = 4.08$ Å), and we assume the muon occupies only octahedral interstitial sites. The volume is taken to be the mean volume per impurity; in our example, this corresponds to a radius of 23.82 Å and includes 51 distinct shells. The first shell at 2.04 Å contains 6 sites, the second at 3.53 Å 8 sites, the third at 4.56 Å 24 sites, etc. The transition probability from the first shell to the second is 1/3, from the first to the third 1/3, from the second to the first 1/4, from the third to the first 1/12. These exhaust all transitions involving the first shell. Similarly, transition probabilities can be determined for all 51 shells. Long-range interactions have a depolarizing effect on all shells, while nearest-neighbor interactions influence the first shell only. We can write a system of differential equations describing the change in polarization for each shell:

$$n_1 dp_1/dt = \tau_\mu^{-1}\sum_{j\neq 1} R_{j1}n_jp_j - \tau_\mu^{-1}\sum_{k\neq 1} R_{1k}n_1p_1 - Cn_1p_1/r_1^6 - Qn_1p_1$$

$$n_2 dp_2/dt = \tau_\mu^{-1}\sum_{j\neq 2} R_{j2}n_jp_j - \tau_\mu^{-1}\sum_{k\neq 2} R_{2k}n_2p_2 - Cn_2p_2/r_2^6 \quad (1)$$

$$\vdots$$

$$n_M dp_M/dt = \tau_\mu^{-1}\sum_{j\neq M} R_{jM}n_jp_j - \tau_\mu^{-1}\sum_{k\neq M} R_{Mk}n_Mp_M - Cn_Mp_M/r_M^6 ,$$

where n_i is the number of octahedral sites on the ith shell, R_{ij} is the transition probability from the jth shell to the ith shell, τ_μ is the mean hop time, C is the strength of the dipole interaction, Q is the strength of the contact interaction, and the total polarization $P(t) = \sum n_ip_i(t)$. We can rewrite eq. (1) in matrix notation:

$$dp/dt = Up \quad . \qquad (2)$$

We assume solutions of the form $p = p_0 \exp(-\lambda t)$, where the λ's are the eigenvalues of the matrix U. To simplify the problem further we have divided shells 4 through 51 into 9 groups and kept the first 3 shells distinct. This approximation is one of convenience and is not strictly necessary; however, it does preserve the correct distances for the innermost shells which experience the greatest depolarization. Of the 12 eigenvalue solutions, λ_i, of eq. (2), only one is found to be appropriate to the time scale of our measurements (microseconds) while the other 11 are at least two orders of magnitude faster. Thus, an exponentially decreasing polarization is predicted and, indeed, observed.

The interaction constants are treated in detail in ref. [illegible]; we simply state them here.

$$C = \frac{1}{15}(\hbar\gamma_s\gamma_\mu)^2 S(S+1)\tau_c\left[4 + \frac{3}{1+\omega_\mu^2\tau_c^2} + \frac{6}{1+\omega_s^2\tau_c^2} + \frac{1}{1+(\omega_s-\omega_\mu)^2\tau_c^2} + \frac{6}{1+(\omega_s+\omega_\mu)^2\tau_c^2}\right] , \qquad (3)$$

where, for Gd^{+3}, $S = 7/2$, $\gamma_s = 17.6 \times 10^6\ s^{-1}g^{-1}$ and $\gamma_\mu = 8.514 \times 10^4\ s^{-1}g^{-1}$. The correlation time is given in terms of the hop time, τ_H, and the impurity spin fluctuation time, τ_F, as $\tau_c = \tau_F\tau_H/(\tau_F + \tau_H)$, where $\tau_F = (\gamma_s b_s T)^{-1}$ and $b_s = 7.5/^oK$.[3] The external field, H_o, enters through the frequency terms, $\omega = \gamma H_o$. We assume, for simplicity, an isotropic contact interaction at the innermost shell of the form $\mathcal{H} = A\,\underline{S}\cdot\underline{I}$ and write A in terms of a field as $A = \gamma_\mu H_x/S$. Then

$$Q = \frac{1}{3}\frac{(S+1)}{S}\gamma_\mu^4 H_x^2 \tau_c \left(1 + \frac{1}{1 + (\omega_s - \omega_\mu)^2\tau_c^2}\right). \qquad (4)$$

In Fig. 1, the experimental depolarization rates for an external field $H_o = 80$ Oe are plotted versus temperature together with the results of the model fit. We also show in Fig. 1 the dipolar contribution to λ as determined by this model and, for comparison, that of Richards' model.[2] The following points should be noted regarding the model predictions:

(a) From the fit values of τ_H we may make an Arrhenius plot (Fig. 2), from which we find an activation temperature of 1510 K and an attempt frequency of $1.88 \times 10^{11}\ s^{-1}$. These can be compared with 545 K and $2.85 \times 10^{7}\ s^{-1}$, respectively, for Cu.[4] The diffusion parameters deduced for muons in Au are closer to those for protons in metals than for muons in Cu and may be indicative of above-barrier diffusion.[5] Approximately the same diffusion parameters are obtained if Richards' diffusion model[1] with renormalized dipolar interaction is used.

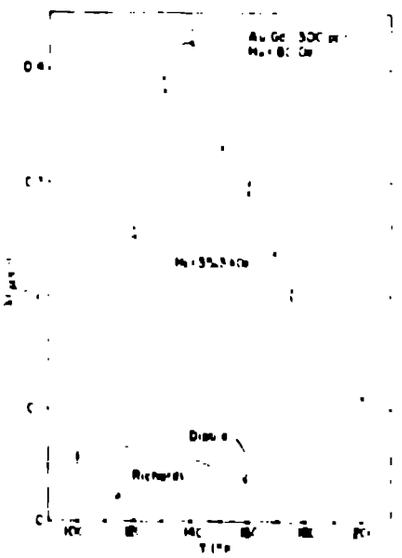

Figure 1. Depolarization rate versus temperature for 80 Oe external field

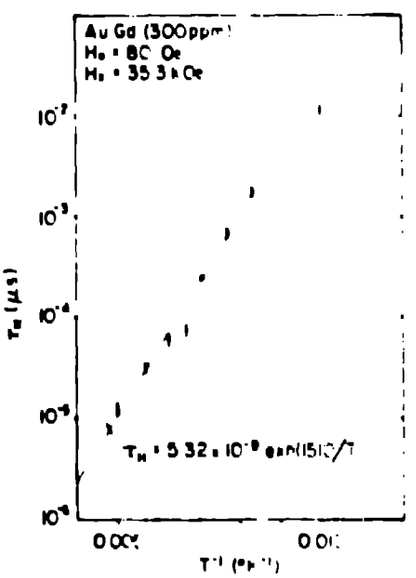

Figure 2. Arrhenius plot with τ_H from data points determined by model.

(b) The contact interaction dominates the dipolar. We find H_x = 35.3 kOe--a reasonable value if the mechanism is, for example, a transferred hyperfine interaction. Such a mechanism was invoked in NMR studies of superionic conductors;[6] however, its suitability in this situation is not obvious and requires further study.

(c) Because the contact interaction involves the innermost shell only, a similar λ vs T curve would be obtained by considering only a dipolar interaction with the innermost shell radius reduced from 2.04 A to 1.32 A. However, it is difficult to believe that the effects of lattice distortion and muon motion would be so dramatic, even considering the strong influence of the r^{-6} factor in eq. (1). (Also, see discussion in ref. 1.)

(d) The dipolar contribution predicted by this model is about three times larger than that predicted by Richards' diffusion model and both are about an order of magnitude smaller than the data.

Figure 3. Depolarization rate versus temperature for 5 kOe external field.

In Fig. 3, we show data taken with the same sample at 5 kOe external field. The fit obtained with all other parameters as in Fig. 1 falls somewhat below the data. To fit this data we must use H_x = 40.7 kOe.

Without a detailed calculation of the contact interaction, it is difficult to account for this discrepancy; however, our assumption of an isotropic contact interaction was made for simplicity and any anisotropy would introduce additional field-dependent terms in eq. (4).

Further refinements of these calculations are planned. These include a proper accounting of the statistical distribution of volumes per impurity and inclusion of tetrahedral interstitial sites for the muon.

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